IV. AIR QUALITY: CHARACTERIZATION AND IMPLICATIONS

This chapter defines the various subclasses of particulate matter (PM) and then briefly discusses the chemical and physical properties of PM in the atmosphere, recent PM concentrations and trends, the relationships between PM and population exposures, and the air quality implications of PM_{10} controls. This information is important both in interpreting the available health effects and welfare information and in making recommendations for appropriate indicators for PM.

A.   Characterization of U.S. Ambient Particulate Matter

PM represents a broad class of chemically and physically diverse substances. The principal common feature of PM is existence as discrete particles in the condensed (liquid or solid) phase spanning several orders of magnitude in size, from molecular clusters of 0.005 µm in diameter to coarse particles on the order of 100 µm.[1] In addition to characterizations by size, particles can be described by their formation mechanism or origin, chemical composition, physical properties, and in terms of what is measured by a particular sampling technique.

In most locations, a variety of diverse activities contribute significantly to PM concentrations, including fuel combustion (from vehicles, power generation, and industrial facilities), residential fireplaces, agricultural and silvicultural burning, and atmospheric formation from gaseous precursors (largely produced from fuel combustion). Other sources include construction and demolition activities, wind blown dust, and road dust. From these diverse sources come the mix of substances that comprise PM. The major chemical constituents of PM_{10} are sulfates, nitrates, carbonaceous compounds (both elemental and organic carbon compounds), acids, ammonium ions, metal compounds, water, and crustal materials. The amounts of these components vary from place to place and over time.

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[1] In this Staff Paper, particle size or diameter refers to aerodynamic diameter, which is defined as the diameter of a spherical particle with equal settling velocity but a material density of 1 g/cm^3, normalizing particles of different shapes and densities (CD, page 3-8).

1.   Multi-modal Size Distributions

The health and environmental effects of PM are strongly related to the size of the particles. The aerodynamic size and associated composition of particles determines their behavior in the respiratory system (i.e., how far the particles are able to penetrate, where particles are deposited, and how effective the body's clearance mechanisms are in removing them as discussed in Chapter V). Furthermore, particle size is one of the most important parameters in determining atmospheric lifetime of particles, which is a key consideration in assessing health effects information because of its relationship to exposure. The total surface area and number of particles, chemical composition, water solubility, formation process, and emission sources all vary with particle size. Particle size is also a determinant of visibility impairment, a welfare consideration linked to fine particle concentrations. Thus, size is an important parameter in characterizing PM, and particle diameter has been used to define the present standards.

The multi-modal distribution of particles based on diameter has long been recognized (Whitby et al., 1972; Whitby et al., 1975; Willeke and Whitby, 1975; National Research Council, 1979; U.S. EPA, 1982a; U.S. EPA, 1982b; U.S. EPA, 1986b; CD Section 3.1.3.2). Although particles display a consistent multi-modal distribution over several physical metrics such as volume and mass, specific distributions may vary over place, conditions, and time because of different sources, atmospheric conditions, and topography. Based on particle size and formation mechanism, particles can be classified into two fundamental modes: fine and coarse modes. Figure IV-1 illustrates an idealized mass distribution of the fine and coarse modes. A depiction of typical number, surface area, and volume distribution of ambient particles is shown in Figure IV-2. This latter figure illustrates that fine particles can be further subdivided into nuclei or ultrafine, and accumulation modes.[2] As illustrated in the figure, even when the fine mode contains about 40

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[2] Typically, the accumulation mode can be characterized by mass median aerodynamic diameter (MMAD) of 0.3 to 0.7 µm and a geometric standard deviation (sigma-g) of 1.5 - 1.8 (CD, page 13-5). The CD defines ultrafine particles as $\leq$ 0.1 µm in diameter (CD, Sections 3.1.3 and 13.2.1). Nuclei or ultrafine particles tend to exist as disaggregated particles for very short periods of time (minutes) and rapidly coagulate into accumulation mode particles (CD page 3-10). Accumulation mode particles, however, do not grow further into the coarse particle mode.

percent of the volume or mass of PM_{10}, it accounts for most of the surface area and number of particles.

The CD concludes that an appropriate cut point[3] for distinguishing between the fine and coarse modes lies in the range of 1.0 µm to 3.0 µm where the minimum mass occurs between the two modes (CD, Section 3.1.2; Miller et al., 1979). The CD states that the data do not provide a clear choice of cut point given the overlap that occurs between the modes. Most ambient measurements of fine particle mass in the U.S. have used instruments with cut points of 2.5 or 2.1 µm. Appendix A outlines the policy considerations involved in making the staff recommendation for using 2.5 µm as the cut point for measuring fine particles.

Table IV-1 introduces some of the size-related terminology used in this Staff Paper. For the purposes of this document, PM_X (e.g., X = 1, 2.5, 10, 15, 10-2.5) is used to refer to gravimetric measurements with a 50 percent cut point of X µm diameter while the terms *fine* or *coarse particles* will be used more generally to refer to the fine and coarse modes of the particle distribution. The distinction highlights the role of formation mechanism and chemistry in addition to size in defining fine and coarse mode particles. Any specific measurement (e.g., $PM_{2.5}$) is only an approximation for fine particles.[4]

In addition to gravimetric fine particle measurements, PM has been characterized in the U.S. and abroad using a variety of filter-based optical techniques including British or black smoke (BS), coefficient of haze (COH), and carbonaceous material (KM), as well as estimates derived from visibility measurements (CD, Chapter 4 and 12; see Appendix B of Staff Paper for limitations in determining mass). In locations where they are calibrated to standard mass units (e.g. London), these measurements can be useful as surrogates for fine particle mass (CD, Chapter 4).

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[3] When used in the context of sampling, *cut point* is a term used to describe the separation efficiency curve for samplers. The cut point is typically described by the aerodynamic diameter at which the sampler achieves 50 percent collection efficiency.

[4] Monitor design, measurement temperature, and inlet efficiency can also affect which particles are included in the definitions of the various size fractions (CD, Chapter 4). Sampling protocols may also affect the amount of semivolatile organics and nitrates and particle-bound water included in a measurement.

The distinction between any specific measurement of fine particles and fine mode (or a measurement of coarse particles and coarse mode) is important because in the subsequent chapters of this Staff Paper, the staff draws public health conclusions regarding fine and coarse mode particles and in doing so the staff relies on the available measurements. Examples of fine particle measurements include $PM_{2.5}$, BS, COH, and concentrations of specific chemical classes predominantly in the fine fraction such as sulfates and acids all judged to be surrogates for fine mode particles. Measurements of coarse particles include $PM_{10-2.5}$, $PM_{15-2.5}$, and TSP minus PM_{10}.

2.   Properties of Fine and Coarse Fraction Particles

As summarized in Table IV-2, fine and coarse particles can be differentiated by their sources and formation processes, chemical composition, solubility, acidity, atmospheric lifetime and behavior, and transport distances (CD Chapter 3). The key properties of fine and coarse particles are described below.

a.   Sources and Formation Processes

Fine and coarse particles generally have distinct sources and formation mechanisms although there may be some overlap. Primary fine particles are formed from condensation of high temperature vapors during combustion (CD, page 3-2). Fine particles are usually formed from gases in three ways: (1) nucleation (i.e., gas molecules coming together to form a new particle), (2) condensation of gases onto existing particles, and (3) by reaction in the liquid phase (CD, page 13-7). Particles formed from nucleation also coagulate to form relatively larger particles, although such particles normally do not grow into the coarse mode (CD, Section 3.1.3.2). Particles formed as a result of chemical reaction of gases in the atmosphere are termed secondary particles because the direct emission from a source is a gas that is subsequently converted to a product that either has a low enough vapor pressure to form a particle or reacts further to form a low vapor pressure substance. Some examples include the conversion of sulfur dioxide (SO_2) to sulfuric acid droplets that further react with ammonium to form particulate sulfate, or the conversion of nitrogen dioxide (NO_2) to nitric acid which reacts further with ammonia to form particulate ammonium nitrate (NH_4NO_3) (CD, Section 3.2.2). Although directly emitted particles are found in the fine fraction (the most common being particles less than 1.0 µm in diameter from combustion sources), particles formed secondarily from gases dominate the fine fraction.

By contrast, most of the coarse fraction particles are emitted directly as particles and result from mechanical disruption such as crushing, grinding, evaporation of sprays, or suspensions of dust from construction and agricultural operations. Simply put, most coarse particles are formed by breaking up bigger particles into smaller ones. Energy considerations normally limit coarse particle sizes to greater than 1.0 µm in diameter (CD, Chapter 3). Some combustion-generated particles such as fly ash are also found in the coarse fraction.

b. Chemical Composition, Solubility, and Acidity

Fine and coarse mode particles generally have distinct chemical composition, solubility, and acidity. Fine mode PM is mainly composed of varying proportions of several major components: sulfates, nitrates, acids, ammonium, elemental carbon, organic carbon compounds, trace elements such as metals, and water. By contrast, coarse fraction constituents are primarily crustal, consisting of Si, Al, Fe, and K (note that small amounts of Fe and K are also found among the fine mode particles but stem from different sources). Biological material such as bacteria, pollen, and spores may also be found in the coarse mode. As a result of the fundamentally different chemical compositions and sources of fine and coarse fraction particles, the chemical composition of the sum of these two fractions, PM_{10}, is more heterogenous than either mode alone.

Figure IV-3 presents a synthesis of the available published data on the chemical composition of $PM_{2.5}$ and coarse fraction particles in U.S. cities by region described in Chapter 6 of the CD. The CD concludes that the fine and coarse fraction are composed of different chemical constituents and that each fraction also has regional patterns resulting from the differences in sources and atmospheric conditions (CD, Section 6.6). Differences across the country in sources and atmospheric conditions contribute to the variability. In addition to the larger relative shares of crustal materials in the West, total concentrations of coarse fraction particles are generally higher in the arid areas of the Western and Southwestern U.S.

In general, fine and coarse particles exhibit different degrees of solubility and acidity. With the exception of carbon and some organic compounds, fine particle mass is largely soluble in water and hygroscopic (i.e., fine particles readily take up and retain water). The fine particle

mode also contains the acidic fraction (CD, Section 3.3.1). By contrast, coarse particles are mostly insoluble, non-hygroscopic, and generally basic.

c.   Atmospheric Behavior

Fine and coarse particles typically exhibit different behavior in the atmosphere. These differences affect several exposure considerations including the representativeness of central-site monitored values and the behavior of particles formed outdoors once inside homes and buildings where people spend most of their time (as discussed below in Section C).

Fine accumulation mode particles typically have longer atmospheric lifetimes (i.e., days to weeks) than coarse particles and tend to be more uniformly dispersed across an urban area or large geographic region, especially in the Eastern U.S. (CD Sections 3.7, 6.3, and 6.4; Wilson et al., 1995; Eldred and Cahill, 1994; Wolff et al., 1985; Shaw and Paur 1983; Altshuller 1982; Leaderer et al., 1982). As noted above, secondary fine particles are formed by atmospheric transformation of gases to particles. Such atmospheric transformation can take place locally during atmospheric stagnation or during transport over long distances. For example, the formation of sulfates from SO_2 emitted by power plants with tall stacks can occur over distances exceeding 300 kilometers and 12 hours of transport time; therefore, the resulting particles are well mixed in the air shed (CD, Sections 3.4.2.1, and 6.4.1) Once formed, the very low dry deposition velocities of fine particles contribute to their persistence and uniformity throughout an air mass (CD, Sections 6.4 and page 7.2; Suh et al., 1995; Burton et al., 1996).

Larger particles generally deposit more rapidly than small particles; as a result, total coarse particle mass will be less uniform in concentration across an urban area than are fine particles (CD, Sections 3.7, and 13.2.4). Because coarse particles may vary in size from about 1 um to over 100 um, it is important to note their wide range of atmospheric behavior characteristics. For example, the larger coarse particles (>10 um) tend to rapidly fall out of the air and have atmospheric lifetimes of only minutes to hours depending on their size and other factors (Wilson and Suh, 1995; Chow et al., 1991; CD, Section 3.2.4). Their spatial impact is typically limited by a tendency to fallout in the proximate area downwind of their emission point. Such large coarse particles are not readily transported across urban or broader areas, because they are generally too large to follow air streams and they tend to be easily removed by impaction on

surfaces (DRI, 1995; CD, Sections 7.2.2 and 13.2.4). The atmospheric behavior of smaller "coarse fraction" particles ($PM_{10-2.5}$) is intermediate between that of the larger coarse particles and smaller fine particles. Thus, coarse fraction particles may have lifetimes on the order of days and travel distances of up to 100 km or more.[5] While it may be reasonable to expect that coarse fraction particles would be less homogeneously distributed across an urban area than fine particles in areas with regionally high fine particle concentrations (e.g. the eastern U.S.), this is not consistently true in a variety of locations (DRI, 1995). In some locations, source distribution and meteorology affects the relative homogeneity of fine and coarse particles, and in some cases, the greater measurement error in estimating coarse fraction mass (Rodes and Evans, 1985) precludes clear conclusions about relative homogeneity.

Nevertheless, because fine particles remain suspended for longer times (typically on the order of days to weeks as opposed to days for coarse fraction particles) and travel much farther (i.e., hundreds to thousands of kilometers) than coarse fraction particles (i.e., tens to hundreds of kilometers), all else being equal, fine particles are theoretically likely to be more uniformly dispersed across urban and regional scales than coarse fraction particles. In contrast, coarse particles tend to be less evenly dispersed around urban areas and exhibit more localized elevated concentrations near sources (CD, Section 13.2.7; DRI, 1995).

d.   Correlations between $PM_{2.5}$ and Coarse Fraction Mass

As might be expected from the differences in origin, composition, and behavior, ambient daily fine and coarse fraction mass concentrations generally are not well correlated. An analysis (SAI, 1996) of several data sets conducted for this review reported the R-squared statistic between daily $PM_{2.5}$ and $PM_{10-2.5}$ mass to be 0.13 for all non-rural sites and 0.21 when rural sites were included.[6] The results indicate a poor correlation between daily averages of the fine and the

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[5] In extreme cases, dust storms occasionally cause very long-range transport of the smaller size coarse particles.

[6] SAI (1996) reported the following:
(1) $R^2 = 0.13$ of daily $PM_{2.5}$ with daily coarse fraction mass concentrations (n = 8,676) between 1988 and 1993 using the Aerometric Information Retrieval System (AIRS), Interagency Monitoring of Protected Visual Environments (IMPROVE), California Air Resources Board (CARB) Dichotomous Network (1990-1993 data), with rural sites removed.
(2) $R^2 = 0.21$ of daily $PM_{2.5}$ with daily coarse fraction mass concentrations (n = 31,510; 57% rural data) between 1985

coarse fractions. In some specific instances, however, fine and coarse fractions may be correlated. For example, a vehicle moving on a dusty road would emit fine particles from the exhaust and produce coarse particle emissions from the road dust. In locations with poorly controlled industrial emissions of both fine and coarse particles, R^2 as high as 0.7 have been reported (Schwartz et al., 1996a).

        e.   <u>Summary</u>

In summary, the fine and coarse mode particles are distinct entities with differing sources and formation processes, chemical composition, atmospheric lifetimes and behaviors, and transport distances. The CD concludes that these profound differences alone justify consideration of fine and coarse fraction particles as separate pollutants for measurement and development of control strategies. The fundamental differences between fine and coarse particles are also important considerations in assessing the available health effects and exposure information.

B.   <u>PM Air Quality Patterns</u>

This section outlines geographic distributions of PM as well as ambient concentration trends and background levels for PM_{10} and fine particles.

1.   PM Concentrations and Trends

        a.   <u>PM_{10} Concentrations and Trends</u>

State and local air pollution control agencies have been collecting PM_{10} mass concentration data using EPA-approved reference samplers and reporting these data to EPA's publicly available AIRS database since mid-1987. Figure IV-4 shows geographic distribution of the 83 areas that are listed as not attaining the current PM_{10} standards as of September 1994; the figure also summarizes the prevalent contributing sources and size of population residing in nonattainment areas. Most of the non-attainment areas are in the Western U.S. with fewer in heavily populated or industrialized eastern areas. Many of the highest values occur in western areas with fugitive dust sources and in mountain valleys impacted by wood smoke during winter inversions (CD, Section 6.5).

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and 1993 using AIRS, IMPROVE, CARB Dichot Network (1990-1993 data), and South Coast Air Basin (SCAB) Intensive Monitoring Network (IMN) (1985-1986).

National trends may readily be examined for the 6-year period from 1988 to 1993 as illustrated in Figures IV-5a and IV-5b. The figures represent 799 trend sites, mostly from urban and suburban locations as well as a few remote locations; monitoring sites with data in at least five of the six years are included. The figures show the trend and site-to-site variability in the composite annual mean and the ninetieth percentile of 24-hour PM_{10} concentrations.[7] The trend for the composite annual mean shows a steady decline totaling 20 percent over the six-year period from 1988 to 1993. The ninetieth percentile similarly decreases 19 percent over the same period (U.S. EPA, 1994a).   Annual average PM_{10} concentrations ranged from 25 to 35 $\mu g/m^3$ for most U.S. regions by 1994. Additional information about current PM_{10} concentrations are presented in Appendix C.

 b. Fine Particle Concentrations and Trends

The $PM_{2.5}$ concentration data are considerably more limited than for PM_{10}. From 1983 to 1993, fewer than 50 sites reported data to AIRS in any given year.[8] Figure IV-6 displays a quarterly smoothed geographic distribution of the IMPROVE and Northeast States Coordinated Air Use Management (NESCAUM) networks' $PM_{2.5}$ data. These data generally do not include urban concentrations but represent the regional non-urban concentrations. The figure shows both the regional character of elevated fine particle levels in the Eastern U.S. and California as well as a strong seasonality. In the Eastern U.S. high fine particle levels dominated by sulfates occur in the summer often in conjunction with elevated ozone levels.

National $PM_{2.5}$ trends are not available because of the limited number of sites measuring $PM_{2.5}$ and the sampling period at most sites is restricted to a few years. The development of national trends is further hindered because $PM_{2.5}$ is measured using a variety of sampling

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[7] The ninetieth percentile statistic is used because PM_{10} sampling frequency varies among sites and may change from one year to the next at some sites. This statistic is less sensitive to changes in sampling frequency than are the maximum or second maximum peak values. Most PM_{10} sites sample on a once every six day schedule.

[8] Additional special studies have also monitored PM2.5, but these data are not reported in AIRS. For this review, EPA assembled other available data sets for analysis (see CD, Section 6.10 and SAI, 1996). The databases assembled to support this Staff Paper include AIRS, Inhalable Particle Network (IPN) (1982-1984), IMPROVE (1987-1995), CARB Dichotomous Network (1990-1993), and SCAB IMN (1985-1986). Figure C-4 in Appendix C provides a summary of the available data for fine particles.

frequencies and a variety of non-standard sampling equipment (because there is currently no federal reference and equivalency program for $PM_{2.5}$).

However, visibility data can be used as a reasonable surrogate to estimate fine particle trends because the extinction coefficient (B_{ext}) is directly related to fine particle mass (CD, page 6-216). Sufficient visibility data are available to produce national trends from 137 U.S. sites (principally airports) since 1948 (CD, Section 6.10.2; NAPAP, 1991). The location of these sites reflects suburban and urban locations with airports. Figure IV-7 depicts trends maps for the 75th percentile extinction coefficient for summer and winter quarters. The figures show significant regional and seasonal trends. In the northeastern states, winter haze shows a 25 percent decrease while in the southeastern states, there is a 40 percent increase in winter haze (NAPAP, 1991).[9] The summer haziness in the Northeast shows an increase up to the mid-1970s followed by a decline. In the Southeast, there was an 80 percent increase in summer haziness, mainly occurring in the 1950s and 1960s (NAPAP, 1991). During the summer months, haziness (extinction coefficient) in the East can be dominated by sulfate (with associated water and ammonium). In this situation, visibility trends may be a better surrogate for sulfate than for non-sulfate related fine particle components (see subsection c below).

Visibility and fine particles have been monitored with more precision by the IMPROVE network from 1987 to present. In eastern remote locations, air quality data from 1982 to 1992 showed roughly a 3 percent annual increase in sulfate mass concentration during the summer and a smaller negative (although not statistically significant) trend in the winter (Eldred and Cahill 1994). Western visibility monitoring through the IMPROVE network has not shown any trends for the period.

c.   Trends in Emissions of Fine Particle Precursor Gases

SO_2, nitrogen oxides (NO_x), which encompasses NO and NO_2, and certain organic compounds are major precursors of secondarily formed fine particles, as described above. The relationship between precursor emission reductions and ambient $PM_{2.5}$ is nonlinear in many

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[9] For the NAPAP analyses, the Northeast was defined as Indiana, Ohio, Pennsylvania, New York, Kentucky, West Virginia and New England states, and the Southeast was defined as states south of the Ohio River and east of the Mississippi (NAPAP, 1991).

aspects; thus, it is difficult to project the impact on $PM_{2.5}$ arising from expected changes in PM precursor emissions without air quality simulation models that incorporate treatment of complex chemical transformation processes. In general terms, one would expect that emission reductions of SO_2 should lead to reductions in sulfate aerosol, but reductions will vary by season, depending on both emission fluctuations and changes in prevailing meteorology and photochemistry.

Figure VI-8 shows comparisons of sulfur emissions for summer and winter with extinction measurements derived from airport visibility data over the Northeast and Southeast in the winter and summer seasons where sulfates are currently the major contributor to light extinction (NAPAP, 1991). The correspondence between sulfur emissions and extinction coefficient is fairly close, particularly in the summer, but not an absolute match. For some years there are increases or decreases in extinction coefficient without corresponding changes in sulfur emissions, which likely reflect changes in non-sulfate particles as well as changes in meteorology and errors in emissions and visibility data. Overall, these data point to a strong relationship between sulfur emissions and regionally occurring fine particle concentrations in the Eastern U.S. (NAPAP, 1991).

It is noteworthy that major reductions in precursor emissions have occurred in the past, such as the large SO_2 reductions that were achieved in the 1970s and 1980s in some locations because of other CAA programs such as the SO_2 NAAQS implementation, prevention of significant deterioration (PSD) program, and later from the new source performance standards (NSPS) program. Similarly, NO_x emissions increases have been limited due to PSD, NSPS, and mobile source control programs. Future reductions in SO_2 of slightly less than 1 percent per year for the next 9 years are projected for the Eastern U.S., primarily from electric utilities (U.S. EPA 1995b). These projected reductions are due to the Acid Deposition Program, as required under Title IV of the 1990 CAA Amendments. Substantial NO_x controls are also required for motor vehicles and utilities under the CAA Amendments.

2.   Background Levels

Natural sources contribute to both fine and coarse particles in the atmosphere. For the purposes of this document, background PM is defined as the distribution of PM concentrations that would be observed in the U.S. in the absence of anthropogenic emissions of PM and

precursor emissions of VOC, NO_x, and SO_x in North America. Estimating background concentrations is important for the health risk analyses presented in Chapter VI and the assessment of fine particle concentrations and visibility effects in Chapter VIII.

Background levels of PM vary by geographic location and season. The natural component of the background arises from physical processes of the atmosphere that entrain small particles of crustal material (i.e., soil) as well as emissions of organic particles and nitrate precursors resulting from natural combustion sources such as wildfire. In addition, certain vegetation can emit fine organic aerosols as well as vapor phase precursors or organic particles. Biogenic sources and volcanos also emit sulfate precursors.   The exact magnitude of this natural portion of PM for a given geographic location can not be precisely determined because it is difficult to distinguish from the long-range transport of anthropogenic particles and precursors.   Based on published reports that attempt to construct a representation of total PM mass from the sum of estimated natural contributions for the PM components noted above, the criteria document provides broad estimates of background PM levels for longer averaging times as shown in Table IV-3.

TABLE IV-3. PM_{10} AND $PM_{2.5}$ REGIONAL BACKGROUND LEVELS

	Western U.S. ($\mu g/m^3$)	Eastern U.S. ($\mu g/m^3$)
PM_{10} , annual average	4 - 8	5 - 11
$PM_{2.5}$, annual average	1 - 4	2 - 5

Source: CD, page 6-44. The lower bounds of the above ranges are based on compilations of natural versus human-made emission levels, ambient measurements in remote areas, and regression studies using human-made and/or natural tracers (NAPAP, 1991; Trijonis, 1982). The upper bounds are derived from the multi-year annual averages of the "clean" remote monitoring sites in the IMPROVE network (Malm et al., 1994). It is important to note, however, that the IMPROVE data used here reflect the effects of background and anthropogenic emissions from within North America and therefore provide conservative estimates of the upper bounds.

As noted in the estimates, there is a definite geographic trend to these levels with the lower values applicable to the Western U.S. and the higher values applicable to the Eastern U.S. The Eastern

U.S. is estimated to have more natural organic fine particles and more water associated with hygroscopic fine particles than the West.

The range of expected background concentrations on a short-term basis is much broader. Specific natural events such as wildfires, volcanic eruptions, and dust storms can lead to very high levels of PM comparable to or greater than those observed in polluted urban atmospheres. Because such excursions are essentially uncontrollable, EPA has developed an "natural events" policy that removes consideration of them from attainment decisions.[10] Disregarding such large and unique events, some estimate of the range of "typical" background on a daily basis can be obtained from reviewing various multi-year data as well as special field studies. On very clean days, IMPROVE daily measurements are less than 1 $\mu g/m^3$ of $PM_{2.5}$. On some days atmospheric conditions are more conducive to accumulation and formation of PM from both natural and anthropogenic emissions sources. Upper bound estimates of daily background as high as 12 $\mu g/m^3$ PM_{10} have been made based on short-duration studies in remote "clean" areas of the Eastern U.S. (Wolff et al., 1983).   Observed peak to mean ratios in natural areas over much longer time periods can provide a rough guide to the highest 24 hour levels arising from "routine" natural emissions and meteorology conducive to maximum particle accumulation. Because such meteorology appears prevalent in the Southeastern US, staff developed 24-hour peak to annual mean ratios for $PM_{2.5}$ data taken from the four Southeastern IMPROVE sites (Bachmann, 1996). If one assumes that the broad regional distribution of anthropogenic and natural sources of PM are somewhat similar, present day observed peak to mean ratios of 2 to 4 can be assumed to apply to the background annual values in Table IV-3. This estimation approach suggests that the highest background 24 hour $PM_{2.5}$ levels over the course of a year could be on the order of 15 to 20 $\mu g/m^3$.

C.   Air Quality Implications for Interpreting Epidemiological Studies

Based on the examination of the substantial body of data, the CD concludes that the differences in exposure relationships alone of fine and coarse fraction particles are sufficient to

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[10]Under the most recent statement (Nichols, 1996), EPA will exercise its discretion not to designate areas as nonattainment and/or to discount data in circumstances where an area would attain but for exceedances that result from uncontrollable natural events. Three categories of natural PM events are specified: volcanic or seismic activity, wildland fires, and high wind dust events.

justify the consideration of fine and coarse particles as separate classes of pollutants (CD page 13-94). The CD notes that the likelihood of ambient fine mode particles being significant contributors to PM-related health effects in sensitive populations (discussed in Chapter V of this Staff Paper) is related to the linkages between fluctuations in outdoor concentrations of PM and personal exposure to outdoor PM, particularly in indoor environments where people spend most of their time and where many chronically ill elderly can be expected to spend all their time (U.S. EPA 1989a; Spengler et al., 1981). In this regard, while both fine and coarse fraction particles can penetrate indoors with similar efficiency (CD, Sections 7.2, 7.7, and 13.2.7; Wallace, 1996; Koutrakis et al., 1992; Lioy et al., 1990), once inside, the longer residence time of fine particles compared to coarse fraction particles enhances the probability of a linkage between fluctuations in outdoor concentrations and day-to-day population exposures for fine mode particles of outdoor origin, as compared to coarse fraction particles of outdoor origin (DRI, 1995; CD, Sections 7.6 and 13.2.7; Wallace, 1996; Anuszewski et al., 1992). In addition, the more uniform distribution of fine particles expected across many urban areas with regionally elevated concentrations and their well-correlated variation from site to site within a given city mean that fine particle measurements at central monitors may provide a better indicator of day-to-day variations in potential exposure to outdoor particles (CD, Section 13.2.7; Burton et al., 1996; Wallace, 1996; Wilson and Suh, 1996).

1.   Representativeness of Central Monitor Measurements of PM Exposures

The CD concludes that central monitoring can be a useful, if imprecise, index for representing the average exposure of people in a community to PM of outdoor origin (CD, Chapter 7; Tamura et al., 1996; Wallace, 1996; Tamura and Ando, 1994; Suh et al., 1993). Thus, for both the prospective cohort and time series epidemiological studies, it appears reasonable to use a representative central monitor or spatially averaged group of monitors to represent the mean community exposure to outdoor PM.

In addition, the CD concludes that fixed-station ambient PM measurements (e.g., PM_{10}, TSP) generally approximate total ambient fine particle exposure more closely than coarse fraction PM exposure (CD Chapter 13.4.3). Within the fine fraction, fixed-station measurements of ambient sulfates likely approximate total exposure to sulfates better than similar measurements of

H^+ characterize total exposure to acidity because a higher proportion of $SO_4^=$ persists indoors (whereas, H^+ is neutralized by indoor ammonia). Thus, the CD concludes that on balance, available health effects estimates from community studies, whatever their magnitude and direction, are subject to more uncertainty for the coarse fraction than the fine mode, and for H^+ than for $SO_4^=$ (CD, page 13-52).

Individual personal exposures to PM can vary considerably from the concentrations measured at a monitoring station. Typically, in the U.S. PM personal exposure measurements are higher than the ambient PM concentrations due to indoor sources of particles such as cooking, smoking, and cleaning. Because of relative day-to-day consistency within any given residence of indoor sources and sinks of PM, the longitudinal (time series) correlation of personal exposure of a specific individual to total indoor PM_{10} (from both outdoor and indoor sources) and ambient PM_{10} can be very high. In homes with minimal indoor sources of PM_{10}, the R^2 values can range above 0.9 when these sources are consistent from day-to-day (CD, page 7-164).

The CD reports similar high correlations between personal and ambient values of sulfate in a cross-sectional exposure study ($R^2 = 0.92$ reported in Suh et al. (1993); CD, page 7-105). Similar high correlations for total sulfur were found by Ozkaynak et al. (1996) in the PTEAM study. These results are noteworthy because unlike $PM_{10,}$ which has both indoor and outdoor sources, sulfate is virtually all of outdoor origin. Consequently, only the traits of the indoor environment, such as air conditioning, modify personal exposures to sulfates while indoors (CD, page 7-105). By contrast, the strength of cross sectional comparisons between total PM_{10} or $PM_{2.5}$ personal exposures and ambient concentrations can vary greatly depending upon the presence of smoking, cooking, or other strong indoor/personal sources (Wallace, 1996).

The day-to-day relationship between PM concentrations monitored at a central station and measurements of personal exposure is important to interpreting the time series community health studies. The CD notes that longitudinal exposure studies are more relevant to interpreting the time series epidemiologic studies than the cross-sectional exposure analyses because the cross-sectional studies often are more influenced by the variations in indoor sources (e.g., one household with a smoker and a smoke-free household) and sinks between subjects (CD, Section 7.4.2; Wallace, 1996). Cross-sectional regression analyses of indoor on outdoor $PM_{2.5}$ and PM_{10}

concentrations generally explain less than half of the variance ($R^2 < 0.50$); however, longitudinal regressions (for a single home measured over a series of days) often have much better indoor-outdoor relationships (R^2 ranging up to 0.9) (CD, Section 7.8).

Thus, the CD concludes that measurements of daily variations of ambient PM concentrations, as used in the time series epidemiologic studies presented in Chapter V, have a plausible linkage to the daily variations of human exposures to PM from ambient sources for the populations represented by the ambient monitoring stations (CD, Chapter 7). The CD concludes that this linkage will be better for indicators of fine particles than for indicators of fine plus coarse particles (i.e., PM_{10} or TSP).

2. $PM_{2.5}$ and PM_{10} Comparisons in Areas Relevant to the Health Studies

Figure IV-9 shows the locations of selected community health studies which reported positive, statistically significant associations between short-term exposure to PM and excess mortality, which are discussed in Chapter V. Significantly, despite the fact that most of the PM_{10} non-attainment areas are mainly in the Western U.S. (see Figure IV-4), the mortality studies were conducted mainly in Eastern U.S. cities, many of which attain the current standards. The eastern sites where studies were conducted have a higher level of regional fine particles (as shown in Figures IV-6 and IV-7). Table IV-4 presents available information about fine particle concentrations in selected cities relevant to the health studies.

By contrast, the coarse fraction in the eastern U.S. is lower, on both an absolute concentration and relative fraction of PM_{10} basis than in the Western U.S. In the Eastern U.S., less than half of the daily PM_{10} mass concentration is coarse fraction material. The seasonal coarse fraction to PM_{10} ratios in the Northeast, for instance, range from 0.36 to 0.38, with an average of all seasons of 0.37 (SAI, 1996).

The Western U.S. has a more complicated pattern of fine and coarse particles because of its more complex mix of sources, topography, and seasonal variability. In some western urban areas, fine particle levels can be equal to or greater than those observed in the Eastern U.S. (see Table IV-4). Urban areas such as Los Angeles, CA, Utah Valley, UT, and Denver, CO, have relatively high contributions of local precursor emissions that may contribute to the formation of fine particles.

D.   Air Quality Implications for Risk Management Strategies

Through the state implementation plan process, State and local agencies are responsible for adopting strategies to control PM in areas with violations of the PM NAAQS.[11] Conversely, areas that currently meet the PM_{10} NAAQS are not required to implement any controls. In non-attainment areas, the implementing agency typically selects control strategies based on its evaluation of which strategies are most effective at reducing PM_{10} concentrations contributing to an exceedance, considering the ability of the area or source to implement the controls and cost. Accordingly, implementing agencies take into account financial costs, availability of technology, suitability of the measure to the specific problem, legal authority of the implementing agency over the emission source (e.g., local sources within a jurisdiction are normally controlled rather than sources of long range transport), and other factors. Because the current standards use a PM_{10} indicator, the extent to which any strategy controls fine or coarse particles is not currently a consideration. As long as the strategies adopted can be reliably demonstrated to provide for expeditious attainment of the standards, EPA does not require one specific measure over another in moderate non-attainment areas. Coarse fraction particles may be preferentially controlled because of their larger contribution to PM_{10} mass concentration in some areas, their local impact, and the relatively lower cost per ton removed.

Of the 83 PM_{10} nonattainment areas shown in Figure IV-4, 37 are eligible for redesignation to attainment, based on air quality data for 1992 to 1994, and an additional seven have preliminary data which suggest they may also be meeting the current standards. The implementation of the PM_{10} NAAQS encompasses diverse sources and solutions. The major sources contributing to PM non-attainment areas include fugitive dust, woodsmoke, stationary sources (e.g., including stacks and materials processing fugitive emissions from steel mills), and mixed areas (that may include the above sources plus additional sources such as regional transport or motor vehicles).

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[11] In moderate non-attainment areas, the CAA requires the application of reasonably available control measures (RACM) and the attainment of the NAAQS as expeditiously as practicable. The expeditiousness test requires the application of reasonably available control technology (RACT). EPA provides guidance on RACM/RACT. Under the guidance, States have flexibility in choosing the mix of controls used to attain the NAAQS.

Table IV-5 presents additional information on the non-attainment areas and the progress towards attainment based on air quality data. Areas dominated by residential woodsmoke and stationary sources have made the most improvement to meet the PM NAAQS, as measured by the number of areas with improved air quality data. Areas with fugitive dust problems and mixed sources (most of which have a fugitive dust problem from activities such as construction and road dust as well as primary and secondary motor vehicle contributions and other sources) have made less progress because local areas with large mobile source contributions have difficulty reducing these emissions and areas with windblown fugitive dust problems are often unable or have limited ability to control the major sources of their problems from soil erosion.

TABLE IV-5. SUMMARY OF PM$_{10}$ NON-ATTAINMENT AREAS BY SOURCE TYPE

Dominant Source Type	Number of PM$_{10}$ Non-attainment Areas	Areas eligible for redesignation based on air quality data*	Difference
Fugitive Dust	23	5	18
Woodsmoke	32	20	12
Stationary Sources	23	12	11
Mixed Sources	5	0	5
Total	83	37	46

* Areas with complete data shown only. Implementing agencies must complete other requirements to be redesignated.

Although implementing agencies have no requirement to consider the relative contributions of fine and coarse particles to the control strategies adopted, national emission inventories and special studies provide some limited information about the relative contributions of fine and coarse fraction particles. Generally, fugitive dust sources tend to produce predominantly coarse fraction particles; residential woodsmoke is predominantly composed of fine particles; and stationary sources typically emit a mixture of fine and coarse fraction particles from a facility (U.S. EPA, 1995b).

Because of the heterogenous nature of the sources of PM$_{10}$, several different types of complex situations confront implementing agencies. Table IV-6 summarizes the relative

contributions of PM_{10} sources and solutions in five areas typical of how successful implementing agencies have dealt with the PM_{10} NAAQS in each of the broader categories described above (Blais, 1996). The additional details in this table make apparent that even in a typical community affected mostly by fine particle residential woodsmoke such as Klamath Falls, OR, as much as 17 percent of the PM_{10} can be attributed to coarse fraction geological material prompting the implementing agency to take appropriate steps to curb these coarse PM_{10} emissions. Some mixed source areas may be able to meet the NAAQS by preferentially controlling the locally emitted coarse fraction particles without controlling fine particles.

The PM NAAQS program has not historically focused on the reduction of PM precursors to reduce PM concentrations except in a few special situations (e.g., Los Angeles, CA, and Provo, UT). Although the CAA requires consideration of secondary PM, implementing agencies are not required to control sources which are not within their non-attainment area or if source-receptor relationships are not established. Many non-attainment areas explicitly do not consider the control of secondary fine PM transported into their area from other sources (e.g., regional background from Ohio River Valley affecting Steubenville, OH, and secondary fine particles from LA Basin affecting Coachella Valley, CA). Instead, implementing agencies preferentially control locally generated coarse and fine fraction sources.